

Photooxidation of Asphalts in the Presence of Ozone

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The effects of ozone on the photooxidation of coating-grade asphalts were compared to those of air and of oxygen under a variety of radiant energy conditions. Oxidative changes were measured by infrared spectroscopy and by the time to film failure as denoted by cracking.

Surfaces of noncatalyzed asphalts were oxidized when treated with ozone in total darkness, room light, solar radiation, and the radiant energies of the carbon and xenon arcs. No oxidation was detected when the asphalts were exposed to room light in oxygen alone. A FeCl_3 -catalyzed asphalt was not oxidized by ozone in total darkness. Photooxidation rates of all asphalts increased as exposures were made to carbon-arc radiation in air, in oxygen, and in ozone-enriched oxygen. Each asphalt exhibited a different oxidative response to solar, xenon-arc, and carbon-arc radiant energy sources.

1. Introduction

In recent years, the high interest in the problem of atmospheric smog has led to extensive research on atmospheric oxidants other than oxygen and their reactions with organic materials in the presence of sunlight [1].¹ While this problem appears pertinent to air-blown asphalts, which are organic materials and are exposed to the same atmospheric conditions during weathering, it has received little attention in asphalt weathering research. Pfeiffer [2] described the effects of various oxidants (aromatic nitro compounds) on asphalt. Nellensteyn and Steffelaar [3] passed nitrogen dioxide through solutions of asphalt in carbon tetrachloride and observed the formation of a precipitate which contained 25 percent oxygen and 5 percent nitrogen. In a very general way, Brown [4] discussed the effects of ozone on asphalt but presented no experimental data.

As part of a program for studying the environmental conditions which influence asphalt degradation, the effects of an ozone-oxygen atmosphere on the photooxidation of asphalt films (25 μ thick) were examined. Using infrared spectroscopy it was possible to follow oxidative changes in asphalt films due to ozonation in the dark, in room light, in sunlight, and in the radiant energies of the carbon arc and the xenon arc, and to compare the changes so produced with those found in oxygen and in air.

2. Experimental Procedure

2.1. Materials

Three commercial coating-grade roofing asphalts (Nos. 6, 16, and 3) and one experimental asphalt (No. 3X) were used in this study. Asphalt No. 6 was of Southeastern U.S.A. origin, asphalt No. 16 was of Midcontinent U.S.A. origin, and asphalt No. 3 was a California Coastal asphalt which had been

catalyzed with ferric chloride. The experimental asphalt No. 3X was blown in the laboratory from the same asphalt flux as the commercial asphalt 3, but did not contain the FeCl_3 catalyst. The physical properties of these asphalts and other pertinent data are given in table 1.

TABLE 1. *Physical properties of asphalts*

No.	Source	Softening point ¹	Penetration at 25 °C	Flash point (COC) ³	Specific gravity at 25 °C	Durability ²
3	California Coastal (catalyzed)	105	18	252	1.032	days 44
3X	California Coastal (noncatalyzed)	104	18			
6	Talco	110	17	288	1.040	41
16	Illinois-Kansas-Oklahoma	117	15	343	1.008	93

¹ Ring and ball method.

² ASTM D 529-59T, "Tentative recommended practice for accelerated weathering of bituminous materials" (51-9C cycle).

³ Cleveland open cup method.

2.2. Ozone Preparation and Analysis

A quartz mercury-arc ultraviolet generator, Ozone Research and Equipment Corporation (Model O₃Q), was used as the ozone source. This instrument produces ozone concentrations from 0 to 20,000 pphm vol⁻¹ (0 to 200 ppm vol⁻¹). Dry oxygen was used in the preparation of the ozone. Ozone concentrations were determined by the iodometric method, i.e., the quantitative release of iodine from an aqueous solution of potassium iodide and titration of the released iodine with sodium thiosulfate (standardized immediately before use). The ozone concentration in the present experiments was 5200 pphm vol⁻¹.²

2.3. Photooxidation

The method used to measure photooxidation rates was based on changes in the infrared spectra of 25- μ

² Separate experiments showed that the amount of ozone generated by the enclosed carbon- and xenon-arc weatherometers was negligible. The open carbon arc (Atlas Weather-Ometer, Model XW) generated ozone as high as 43 pphm vol⁻¹.

¹ Figures in brackets indicate the literature references at the end of this paper.

thick films at 1700 cm^{-1} ($5.88\text{ }\mu$) due to exposure to carbon-arc radiant energy. Detailed procedures for the preparation, irradiation, and infrared analysis of these films in air have been reported previously [5, 6, 7]. The irradiation of asphalt films in an ozone-oxygen or oxygen atmosphere required modified aluminum holders and glass apparatus designed to permit replacement of air with the selected atmosphere and subsequent irradiation in that environment.

The use of aluminum panels has proved very useful in conjunction with the infrared analysis of asphalt thin films [6]. However, new modified aluminum holders have been designed for use where space is limited. The holder is depicted in figure 1. The holder dimensions are $3/8\text{ in.} \times 1/4\text{ in.} \times 2\text{ in.}$, with an opening $5/16\text{ in.} \times 13/16\text{ in.}$, which is larger than the slit opening of the infrared spectrophotometer. The film holder may then be fitted tightly into a modified spectrophotometer sample holder. The experimental procedure was the same as that used with the regular aluminum panels. Separate experiments established that ΔA values (changes in absorbance at 1700 cm^{-1}) obtained from asphalt films mounted on either the usual aluminum panels or on the newer film holders were identical. However, time to film failure was altered, due to the different dimensions of the unsupported asphalt film. In general, asphalt films mounted on the smaller holders did not crack as soon as those mounted on the larger panels.

The irradiation of thin films of asphalt in an atmosphere other than air necessitated the construction of exposure tubes which would transmit appreciable amounts of radiant energy and which could be conveniently sealed. For this purpose, flat culture tubes (Corning 9200, $23\text{ mm} \times 11.5\text{ mm} \times 152\text{ mm}$) were modified with ball joints and with stopcocks. The tubes used had a range in transmission of 81.5 to 82.05 percent at $350\text{ m}\mu$ as measured by a Beckman DU Spectrophotometer. The assembled apparatus is shown in figure 2.

Most of the asphalt exposures were made in an Atlas Weather-Ometer, Model SMC-R, equipped with an automatic line voltage regulator which

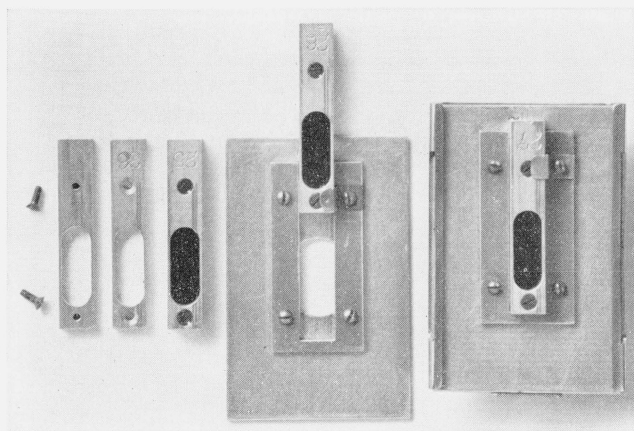


FIGURE 1. Asphalt-film exposure holder.

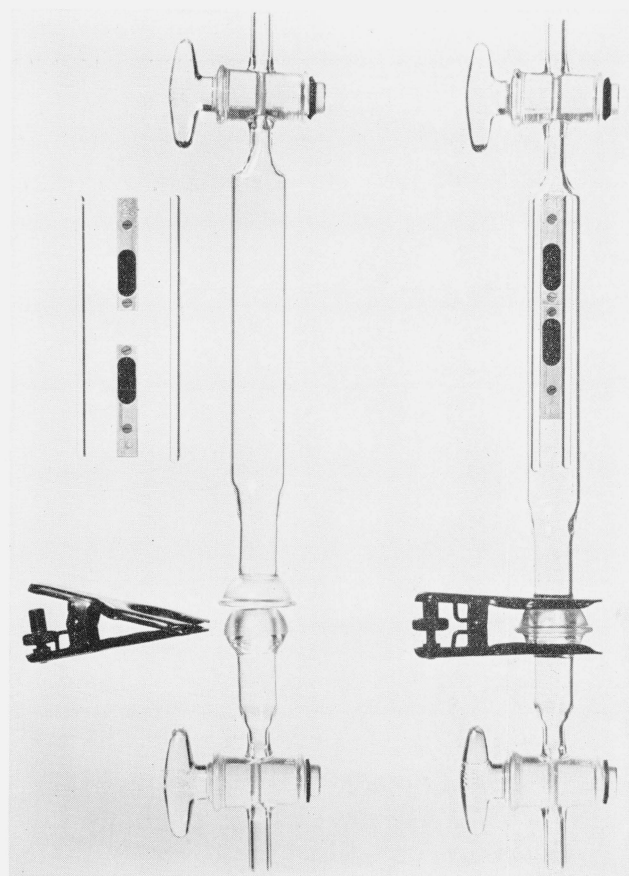


FIGURE 2. Glass irradiation apparatus for asphalt films.

controlled arc voltage at $210\text{ V} \pm 1\text{ percent}$. This machine controlled temperature within $\pm 1\text{ }^{\circ}\text{C}$ of a selected value. The radiant energy source was a single, enclosed carbon arc. Exposures were made at a black panel temperature of $49\text{ }^{\circ}\text{C}$ for selected durations. For those exposures in an ozone-oxygen atmosphere or in oxygen, duplicate specimens were placed in the glass apparatus, flushed for 1 hr (in light and at room temperature), and then irradiated. For comparative purposes, exposures in air were also made in the glass apparatus. The exposures of asphalt in ozone-oxygen to solar radiant energy were made on a clear, sunny day in February in Washington, D.C. The xenon-arc exposures were made at $49\text{ }^{\circ}\text{C}$ with Atlas Weather-Ometer, Model 60-WR, equipped with a constant wattage transformer.

3. Results and Discussion

3.1. Southeastern U.S.A. Asphalt

Films of commercial asphalt 6 were found to undergo oxidation and/or photooxidation under all experimental conditions in which the asphalt surface was brought into contact with ozone. The resultant oxidative changes are shown by the infrared spectra in figure 3a. The spectrum of the original asphalt,

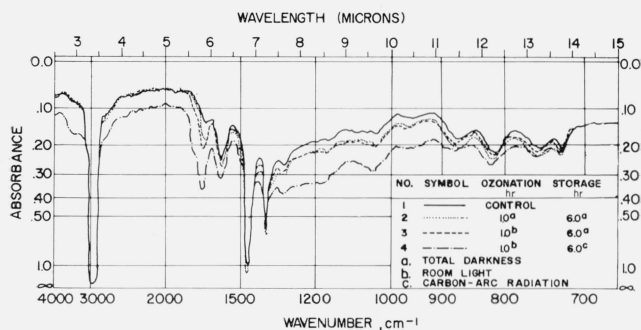


FIGURE 3a. Infrared spectra of asphalt No. 6 showing the effects of ozone under various radiant energy conditions (see text).

which had been exposed to room light and air, is shown in curve 1. The second spectrum (curve 2) is that of asphalt 6 after it had been flushed with ozone for 1.0 hr and stored for 6.0 hr, all in total darkness. Increased absorbance was observed in all of the identifiable oxygen-containing functional groups, the greatest change being in the carbonyl band at 1700 cm^{-1} . The ΔA value at this wavenumber was 0.025 absorbance units.

The effect of room light on the oxidation of asphalt 6 in the presence of ozone is shown in curve 3. The asphalt was flushed for 1.0 hr in room light followed by 6.0 hr storage in the dark. Changes in absorbance as compared to curve 2 were primarily in the carbonyl band where a ΔA value of 0.066 was obtained. The same experiment using oxygen, produced no detectable oxidation.

Far greater effects were found, however, when films of asphalt 6 were flushed with ozone-enriched oxygen for 1.0 hr in room light and exposed to the radiant energy of the carbon arc for 6.0 hr at a black panel temperature of 49°C (curve 4). Pronounced changes were observed in the infrared spectrum from 4000 to 850 cm^{-1} . The ΔA value at 1700 cm^{-1} was 0.172.

Films of asphalt 6 in ozone-oxygen were exposed to two other radiant energy sources, i.e., solar radiant energy and to the xenon arc. Both sources promoted extensive photooxidation over a 6-hr period. Infrared spectra of the exposed asphalt films are shown in figure 3b. The changes in absorbances of the oxygen-containing functional groups were, in general, the same as those produced in the carbon arc. Since the asphalt specimens received different incident radiant energy dosages from each source, absorbances cannot be compared directly. However, the change in the absorbance of any given asphalt exposed to ozone in total darkness may be compared directly with the changes occurring when the same asphalt is exposed to radiant energy sources. In all cases, much more oxidation occurred in the presence of radiant energy.

In a series of experiments designed to observe photooxidation of asphalt in the presence of air, oxygen, and ozone-enriched oxygen under the same conditions of carbon-arc exposure time, intensity, and temperature, the presence of ozone greatly accelerated the photooxidation reaction(s). Oxidative

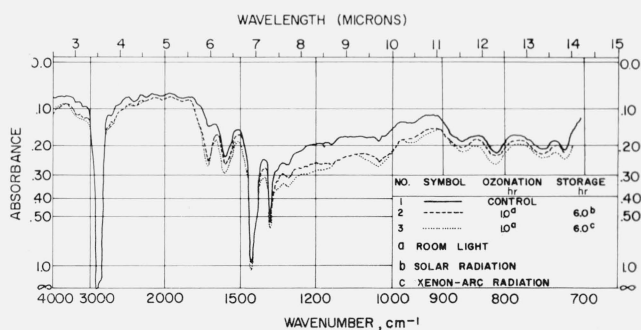


FIGURE 3b. Infrared spectra of asphalt No. 6 showing the effects of ozone in the presence of solar and xenon-arc radiation.

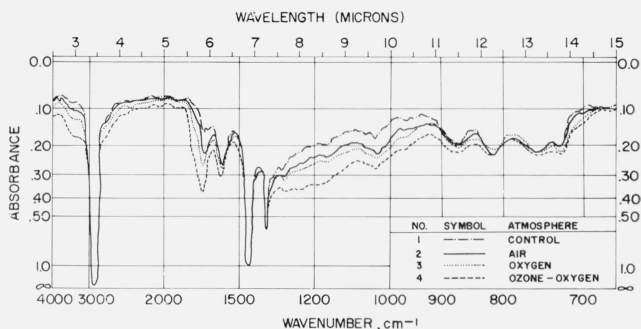


FIGURE 4. Infrared spectra of asphalt No. 6 showing the effects of exposure to the carbon arc (49°C , 6 hr) in various environments.

changes were measured by infrared analysis as in the previous experiments. The spectra of the exposed asphalts are shown in figure 4. The respective ΔA values at 1700 cm^{-1} obtained with air, oxygen, and ozone-enriched oxygen after 6 hr exposure were 0.074, 0.098, and 0.182. Also, in the presence of ozone, large increases in absorbance were observed in other bands of oxygen-containing functional groups, namely, the 3500 cm^{-1} region (alcohols, free OH absorbances), the 2700 cm^{-1} region (acids, bonded OH stretching frequencies), and at 1600 cm^{-1} (aromatic and conjugated alkene and OH bonding). Much less absorbance was observed in these same areas when exposures were made in air or in oxygen.

In photooxidation-rate experiments with commercial asphalt 6 in air, in oxygen, and in ozone-enriched oxygen, respectively, the length of the induction period, the slope of the straight-line portion of the curve, and the time to film failure were affected by the atmosphere used (fig. 5). (Film failure, as denoted by cracking, occurred during the time interval following the last infrared scan and before the next sampling period.) The induction period in oxygen was less than that in air. In the presence of ozone, however, the induction period was completely eliminated. (Although the curve begins at the origin, it should be realized that some oxidation occurred

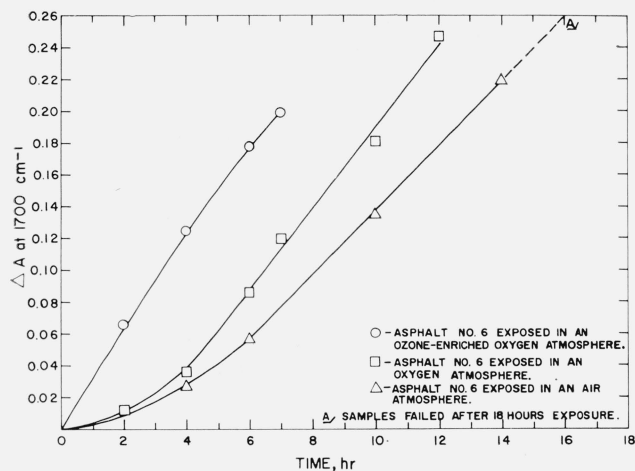


FIGURE 5. Effects of various environments on asphalt No. 6 as measured by changes in infrared absorbance at 1700 cm^{-1} with time of carbon-arc exposure (49°C).

during the ozone-flushing period.) The slopes of the curves are $0.0203\ \Delta A/\text{hr}$ for air, $0.0258\ \Delta A/\text{hr}$ for oxygen, and $0.0308\ \Delta A/\text{hr}$ for ozone-enriched oxygen.³ When the slope for air is considered as unity, the ratio of the slopes for air, oxygen, and ozone-enriched oxygen becomes 1.0: 1.27: 1.52. Overall photodegradation rates as measured by film failure time for air were approximately 18 hr, for oxygen 12 hr, and for ozone-enriched oxygen 7 hr. Treatment of asphalt with ozone caused an approximate three-fold increase in photooxidation.

3.2. Midcontinent U.S.A. Asphalt

Experiments comparable to those described in section 3.1 were made with asphalt No. 16, a Midcontinent U.S.A. asphalt which had a durability (carbon-arc, 51–9C cycle) of 93 days (table 1). Ozonation of this asphalt in total darkness as well as to various radiant energy sources (carbon arc, xenon arc, and sunshine) produced results similar to those with asphalt 6. Changes as detected by infrared spectroscopy are shown in figures 6a and 6b. The greater stability of asphalt 16 to photodegradation is manifested by the smaller absorbance changes for any given radiant energy source. However, even in total darkness oxidation occurred to some extent, particularly in the carbonyl absorbance at 1700 cm^{-1} .

The effect of ozone, for a given radiant energy source (carbon arc), is clearly demonstrated in figure 7 where typical infrared spectra are given for 10-hr exposures in air, oxygen, and ozone-oxygen, respectively. Ozone greatly increased the amount of photooxidation that took place. The average ΔA (1700 cm^{-1}) values for air, oxygen, and ozone-oxygen were 0.062, 0.100, and 0.175, respectively.

³ A straight-line slope was calculated for this system even though the curve deviates slightly from linearity for the last two exposure points.

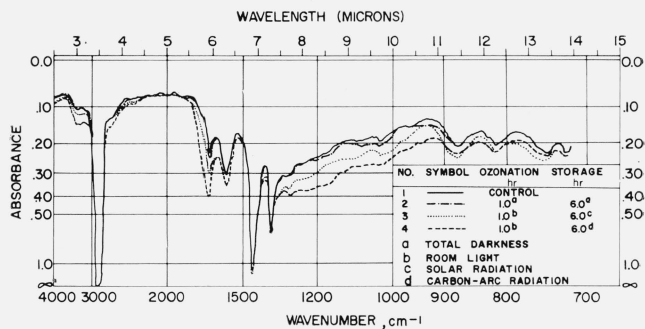


FIGURE 6a. Infrared spectra of asphalt No. 16 showing the effects of ozone under various radiant energy conditions.

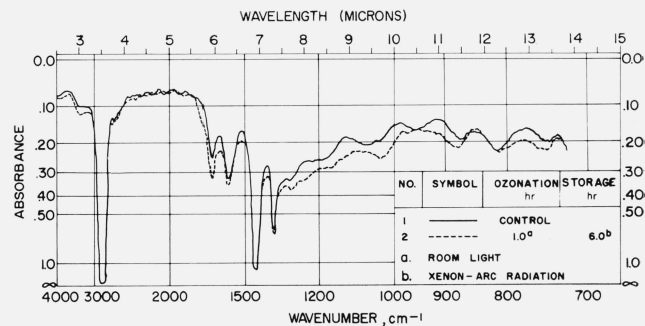


FIGURE 6b. Infrared spectra of asphalt No. 16 showing the effects of ozone in the presence of xenon-arc radiation.

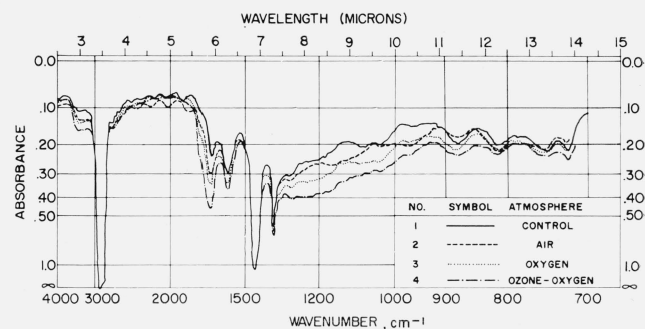


FIGURE 7. Infrared spectra of asphalt No. 16 showing the effects of exposure to the carbon arc (49°C , 10 hr) in various environments.

The accelerating effect of ozone on the photooxidation of asphalt was demonstrated further by photooxidation rate experiments in the three respective gaseous environments. The rates may be compared in figure 8. For direct comparison the exposures in air were made in the glass apparatus as described in section 2.3.

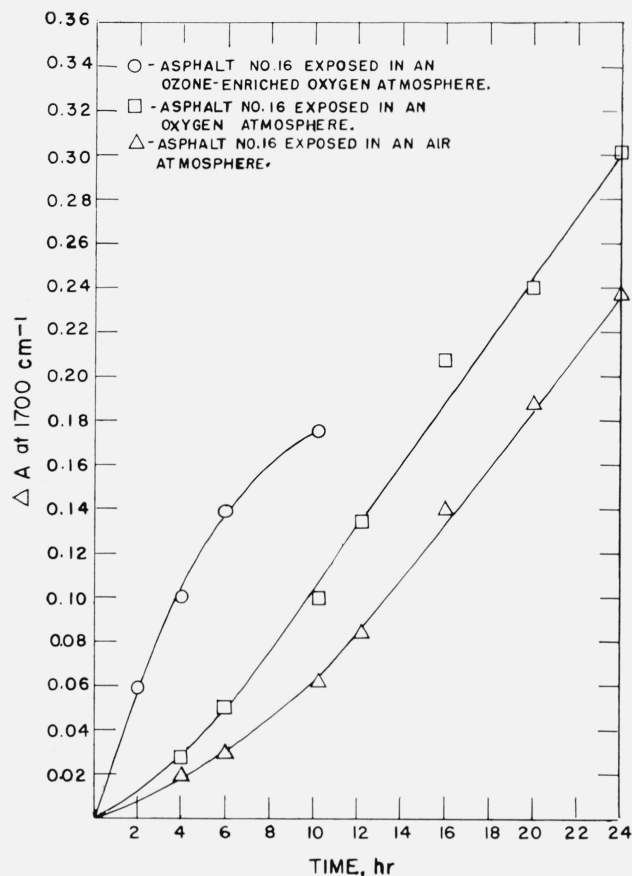


FIGURE 8. Effects of various environments on asphalt No. 16 as measured by changes in infrared absorbance at 1700 cm^{-1} with time of carbon arc exposure (49°C).

The results with asphalt 16 were similar to those of asphalt 6 in that the environments in the order of increasing oxidation rates were air, oxygen, and ozone-oxygen. With both asphalts, ozone reduced the film failure time to approximately one-half that in air. The values of ΔA (1700 cm^{-1}) per hour for the straight-line portions of the curves for asphalt 16 were 0.0126 in air, 0.0145 in oxygen, and 0.0200 in ozone-enriched oxygen. These results again manifest the greater photooxidation stability of asphalt 16 (93 days, 51-9C cycle) over that of asphalt 6 (41 days, 51-9C cycle).

3.3. California Coastal Asphalt

In addition to being from a different geographical source, the California Coastal asphalt (No. 3) differed from asphalts 6 and 16 in that it contained ferric chloride, an air-blowing (asphalt hardening) catalyst. Since this catalyst produced some unusual effects in the asphalt hardening process (necessary to make a coating-grade asphalt) [8], it was included in the present work to elucidate any effects on the photooxidation process with ozone. In order to separate the effects of catalyst and source, some experiments were made with asphalt No. 3X which

had been air-blown in the laboratory from the same asphalt flux as that used for asphalt 3, but without the FeCl_3 catalyst.

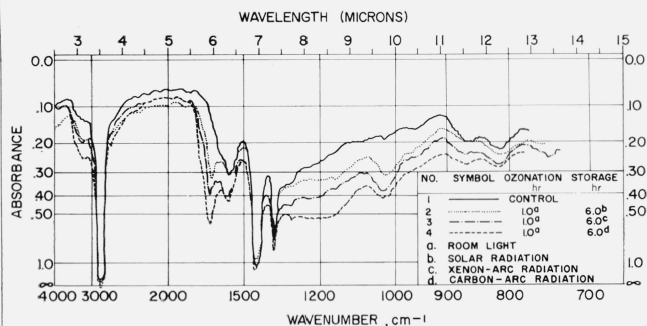
Upon exposure to ozone in total darkness, asphalt 3 showed no detectable oxidation as determined by infrared techniques. Similar treatment of asphalt 3X produced measurable changes in the carbonyl absorbance at 1700 cm^{-1} as had asphalts 6 and 16. Thus, FeCl_3 appeared to inhibit the oxidation reaction caused by ozone in the darkness. (No oxidation could be induced with oxygen alone with any of the asphalts in total darkness.)

In the presence of radiant energy (solar, carbon arc, and xenon arc), however, photooxidation of asphalt 3 took place readily. Changes, as measured by infrared techniques, are shown in figure 9 for 6.0 hr exposures of asphalt 3 in ozone-oxygen to these three radiant energy sources. As with the previous asphalts, absorbance intensities are not directly comparable due to different incident radiant energy dosage for each source.

In general, absorbance changes indicate the formation of the same types of oxygen-containing groups in the three environments. A marked similarity of the spectra in the 4000 to 3000 cm^{-1} region (free hydroxyl) may be observed for the solar and xenon-arc exposures. The greater absorbance change caused by carbon-arc exposure may well be in part an effect of the difference in the spectral distribution of the radiant energy from this source.

Campbell and Wright have reported previously [8] that FeCl_3 appeared to act as a photosensitizer when FeCl_3 -catalyzed asphalt was irradiated (carbon arc) in air. A direct comparison was made in the present work between asphalts 3 and 3X in ozone-oxygen to 6.0 hr solar irradiation. The carbonyl absorbance change, ΔA at 1700 cm^{-1} , was 0.101 for asphalt 3X and 0.133 for asphalt 3. These results confirm those for photooxidation in air and indicate that FeCl_3 may well accelerate the photooxidation of asphalt, but inhibits oxidation in total darkness.

The response of asphalt 3 to the conditions of photooxidation with air, oxygen, and ozone-enriched oxygen are shown in figure 10. The radiant energy source was the carbon arc and an exposure duration



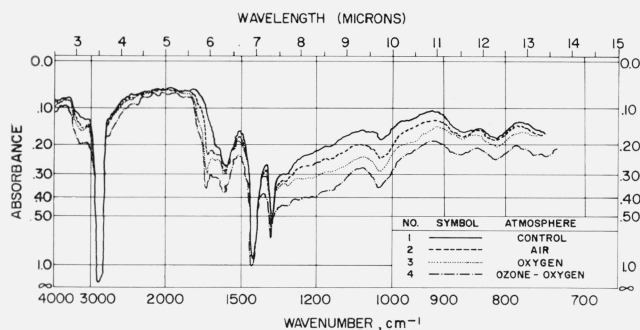


FIGURE 10. Infrared spectra of asphalt No. 3 showing the effects of exposure to the carbon arc (49 °C, 3 hr) in various environments.

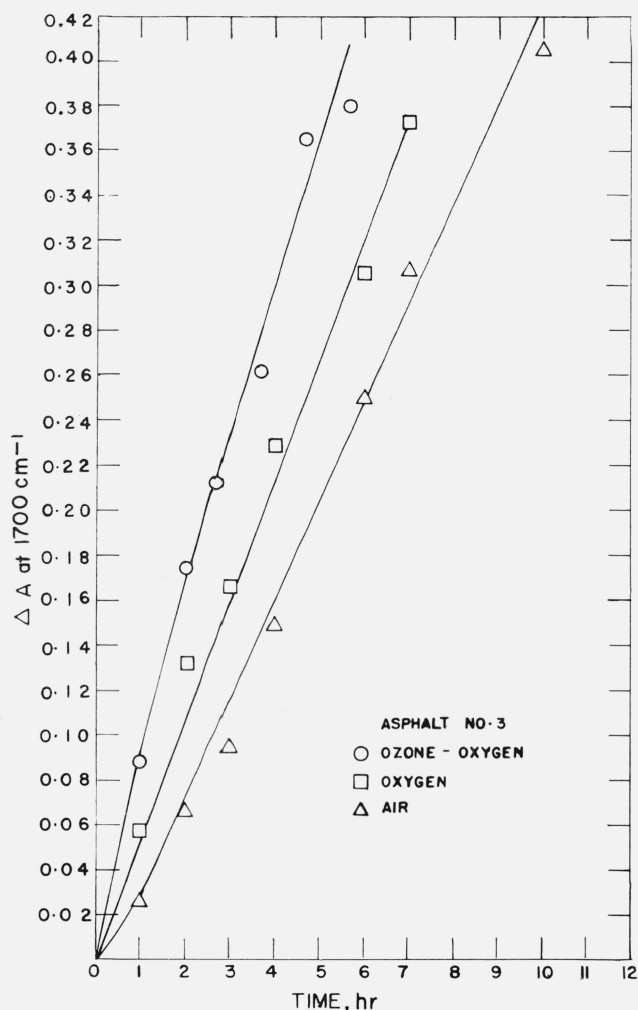


FIGURE 11. Effects of various environments on asphalt No. 3 as measured by changes in infrared absorbance at 1700 cm⁻¹ with time of carbon-arc exposure (49 °C).

of 3 hr was used. The general effect was the same as for asphalts 6 and 16, namely, that the amount of photooxidation increased as the environment was changed from air to oxygen to ozone-enriched oxygen, respectively.

This same effect, i.e., the acceleration of photooxidation with ozone, was confirmed by a series of photooxidation-rate experiments in air, oxygen, and ozone-enriched oxygen as shown in figure 11. The values of ΔA (1700 cm⁻¹) per hour for the straight-line part of the curves produced slopes of 0.0463 in air, 0.0538 in oxygen, and 0.0660 in ozone-oxygen. It may be noted that in both oxygen and ozone-oxygen no induction period was observed.

Asphalts 6, 16, and 3 had more than a twofold range in durability (carbon arc, 51-9C cycle), yet, when photooxidized in ozone, the time to film failure due to cracking was reduced by one-half for each asphalt. As mentioned previously, infrared spectra indicated that the same photooxidation products were formed in air, oxygen, or in ozone. Therefore, ozone could well be used as a realistic means of accelerating the photodegradation of asphalt in laboratory weathering.

3.4. Interrelation of Asphalt, Radiance Sources, and Gaseous Environments

Figures 12a, 12b, and 12c provide a direct comparison of the photooxidation rates for asphalts 6, 16, and 3, in air, oxygen, and ozone-enriched oxygen, respectively. With all three gaseous systems the asphalts in order of decreasing photooxidation stability are California Coastal, Southeastern U.S.A., and Mid continent U.S.A. For air (fig. 12a), the shapes of the curves were similar for each asphalt. With oxygen (fig. 12b), this similarity remained with the exception that the California asphalt had no measurable induction period. The curves for ozone-enriched oxygen (fig. 12c), were different than for oxygen or air in that no induction periods were observed with any of the asphalts. Also, for the two asphalts which failed after 6 hours, a definite decrease in rate occurred. It is possible that such an effect could be caused by total utilization of the ozone after 6-hours exposure time and that ozone, as well as some component of the asphalt, is necessary throughout the photooxidation process. Dardin and Albright [9] have shown that ozone serves to initiate the free radical, thermal, decomposition of propane and is not required throughout the reaction period. While in the present work degradation was promoted by photolysis, the reactions are believed to be the free-radical type [1] and, thus, similar to that described in [9]. However, if the photooxidation rate decreased due to exhaustion of ozone, it should thereafter remain about the same as that found for exposures in oxygen alone. Since this is not the case (figs. 5 and 8), it suggested that a "skinning" effect may be taking place on the asphalt surface and thereby reducing the oxidation rate. Dickinson et al. [10] have reported such an effect in oxygen and in air, but for longer exposure periods.

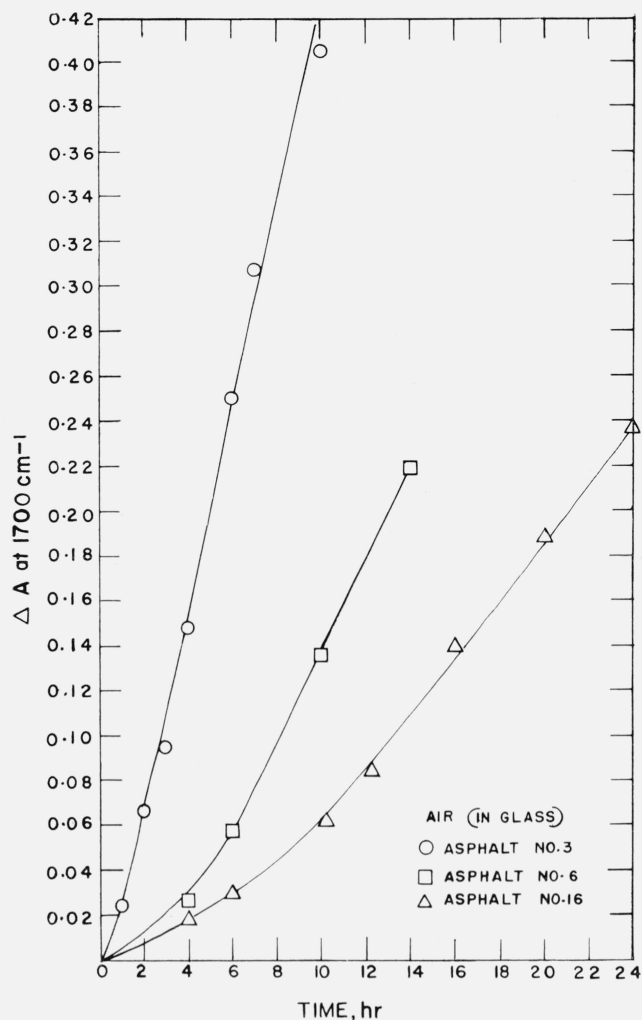


FIGURE 12a. Effects of air on asphalts No. 3, 6, and 16 as measured by changes in infrared absorbance at 1700 cm^{-1} with time of carbon-arc exposure (49°C).

The effects of gaseous environments on the photooxidation rates of asphalts 3, 6, and 16 are compared further in table 2. The slopes of the curves for each asphalt in air, oxygen, and ozone are presented ($\Delta A/\text{hr}$) and then compared on a common basis, namely, the ratios of the rates in oxygen and in ozone to those in air as unity. For all three asphalts, a slight but noticeable increase was noted in the oxidation rate in oxygen compared to that in air. This increase would be expected on the basis of the higher oxygen concentration alone. However, the rates in ozone-oxygen show a pronounced increase over those both in air and in oxygen, indicating considerable more reactivity under photooxidation conditions. Furthermore, the asphalt most durable by the ASTM test, No. 16, had the highest relative rate in ozone, while the least durable, No. 3, had the

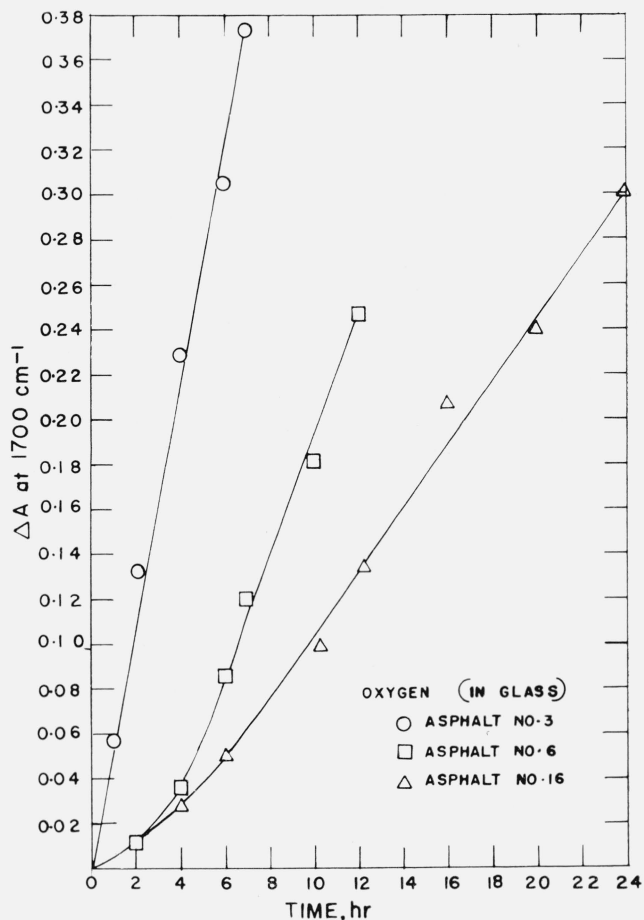


FIGURE 12b. Effects of oxygen on asphalts No. 3, 6, and 16 as measured by changes in infrared absorbance at 1700 cm^{-1} with time of carbon-arc exposure (49°C).

lowest. Thus, it may be inferred that the Mid-continent U.S.A. asphalt, No. 16, had more reactive sites for ozone attack than did the California Coastal asphalt, No. 3.

TABLE 2. Effect of gaseous environment on photooxidation rates ¹ of various asphalts

Asphalt	Photooxidation rates in:			Relative ratio of photooxidation rates ²		
	Air	Oxygen	Oxygen+ozone ³	Air	Oxygen	Oxygen+ozone ³
3	0.0463	0.0538	0.0660	1.0	1.16	1.42
6	.0203	.0258	.0308	1.0	1.27	1.52
16	.0126	.0145	.0200	1.0	1.15	1.59

¹ Expressed as $\Delta A/\text{hr}$. Absorbances measured at 1700 cm^{-1} .

² Rate for air considered as unity.

³ Ozone concentration 5200 ppm vol-1 in oxygen.

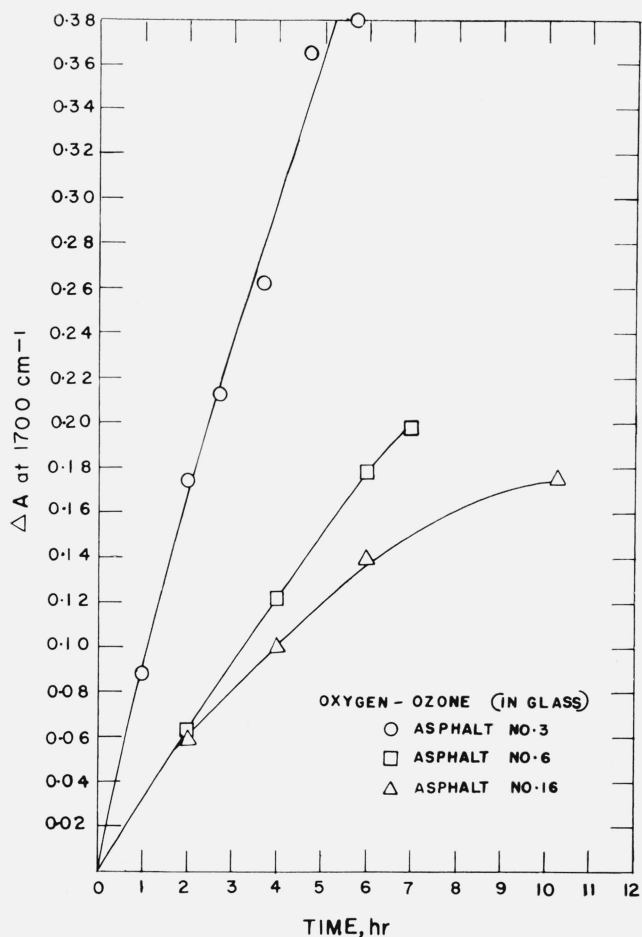


FIGURE 12c. Effects of ozone-enriched oxygen on asphalts No. 3, 6, and 16 as measured by changes in infrared absorbance at 1700 cm^{-1} with time of carbon-arc exposure (49°C).

In table 3, the response of four asphalts (3, 3X, 6, 16) to solar, xenon-arc, and carbon-arc radiant energy are compared. An exposure period of 6 hr was used for each source and asphalt response was measured by ΔA values at 1700 cm^{-1} . For direct comparison, the ratios of ΔA values are shown for xenon-arc, carbon-arc, and solar exposures with the latter

considered as unity. These ratios show that: (a) each asphalt responded differently to each radiant energy source, and (b) as would be expected from the spectral distribution of radiant energy, the response to the xenon source was much more like that to solar exposure than was the response to the carbon arc. These preliminary results are indicative of the selectivity of individual asphalts toward radiant energy sources.

TABLE 3. Ozone-oxygen exposures of four asphalts to various radiance sources ¹

Asphalt	Source of radiance			Relative response ²		
	Solar	Xenon arc	Carbon arc	Solar	Xenon arc	Carbon arc
	ΔA values (1700 cm^{-1})					
3	0.135	0.184	0.408	1	1.36	3.02
3X	.111	.142	-----	1	1.28	-----
6	.091	.093	.178	1	1.02	1.96
16	.040	.066	.139	1	1.65	3.47

¹ Exposed for 6 hr. Ozone concentration in oxygen = 5200 pphm vol-1.

² ΔA for solar radiation considered as unity.

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